

Application Serial No. 09/801,390

Docket No. 2000-022R1  
PATENTAMENDMENTS TO THE SPECIFICATION

Please substitute the following replacement paragraph(s) [0001], [0038], [0121], [0126], [0146] and [0148] for the previously-pending versions of such paragraph(s). Each of these replacement paragraph(s) are amended herein, and marked-up to show changes from the previously-pending versions thereof.

**[0001]** This application is a continuation-in-part of co-owned, co-pending U.S. patent application Ser. No. 09/518,794, entitled "Chemical Processing Microsystems, Diffusion-Mixed Microreactors and Methods for Preparing and Using Same", filed March 3, 2000 by Bergh *et al.*, now issued as U.S. Patent No. 6,749,814. The present invention is also related to, and claims priority to co-owned, co-pending U.S. patent application Ser. No. 60/187,566 entitled "Apparatus and Methods for Multi-Variable Optimization of Reaction Systems and Other Chemical Processing Microsystems", filed March 7, 2000 by Bergh *et al.*, and to co-owned, co-pending U.S. patent application Ser. No. 60/229,984 entitled "Apparatus and Methods for Optimization of Process Variables in Reaction Systems and Other Chemical Processing Systems", filed September 2, 2001 by Bergh *et al.*, ~~each now abandoned in favor of the present application.~~

**[0038]** The present invention is related to the following patents and/or patent applications, each of which is hereby incorporated by reference for all purposes, including for the purpose of combination of various features disclosed in the various related applications to various features disclosed herein, to the highest extent practical, based on the knowledge in the art, and coupled with the guidance of this application and the related applications: (1) co-owned, co-pending U.S. patent application Ser. No. 60/187,566 entitled "Apparatus and Methods for Multi-Variable Optimization of Reaction Systems and Other Chemical Processing Microsystems", filed March 7, 2000 by Bergh *et al.*, ~~now abandoned in favor of the present application;~~ (2) co-owned, co-pending U.S. patent application Ser. No. 60/229,984 entitled "Apparatus and Methods for Optimization of Process Variables in Reaction Systems and Other Chemical Processing Systems", filed September 2, 2001 by Bergh *et al.*, ~~now abandoned in favor of the present application;~~ (3) co-owned, co-pending U.S. patent application Ser. No. 09/093,870, entitled "Parallel Fixed-Bed Reactor and Fluid Contacting Apparatus and Method", filed June 9, 1998 by

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Guan *et al.*, and now issued as U.S. Patent No. 6,149,882; (4) to co-owned, co-pending U.S. patent application Ser. No. 09/518,794, entitled "Chemical Processing Microsystems, Diffusion-Mixed Microreactors and Methods for Preparing and Using Same", filed March 3, 2000 by Bergh *et al.*, issued as U.S. Patent No. 6,749,814; (5) U.S. Serial No. 60/274,065, entitled "Parallel Flow Reactor Having Improved Thermal Control" filed on March 7, 2001 by Bergh *et al.*, ~~abandoned in favor of~~ filed as non-provisional application U.S. Ser. No. 10/094,257 (pending and published as US 2002-0170976); (6) U.S. Serial No. 60/274,022, entitled "Gas Chromatograph Injection Valve Having Microvalve Array" filed on March 7, 2001 by Bergh *et al.*, ~~abandoned in favor of~~ later filed as non-provisional applications U.S. Ser. No. 10/092,364 (pending and published as US 2002-0127146) and ~~in favor of~~ U.S. Ser. No. 10/092,035 (issued as U.S. Patent No. 6,742,544); and (7) U.S. Serial No. 09/801,430 entitled "Parallel Gas Chromatograph With Microdetector Array" filed on March 7, 2001 by Srinivasan *et al.* (issued as U.S. Patent No. 6,701,774). Further reference to several of these applications is made below, in the context of the present invention.

[0121] Particularly preferred parallel flow reaction systems can include a reactor module that comprises four or more parallel flow reactors having separate and independent temperature control for each of the four or more reactors. Substantial thermal management challenges exist for such reactor modules in which the four or more flow reactors are close-packed – that is, in which the four or more reactors have a spatial density (taken along one or more cross-sections) of not less than about 1 reactor / 100 cm<sup>2</sup>, preferably not less than about 1 reactor / 50 cm<sup>2</sup>, more preferably not less than about 1 reactor / 10 cm<sup>2</sup>, and, in some applications, not less than about 1 reactor / cm<sup>2</sup>, not less than about 2 reactors / cm<sup>2</sup>, not less than about 1 reactor / mm<sup>2</sup>. A preferred approach for establishing thermal independence between each of the four or more reactors, such that simultaneous independent temperature control can be effected for each of the four or more reactors without substantial thermal interference from adjacent reactors is disclosed in U.S. Serial No. 60/274,065, entitled "Parallel Flow Reactor Having Improved Thermal Control" filed on March 7, 2001 by Bergh *et al.*, ~~abandoned in favor of~~ filed as non-provisional application U.S. Ser. No. 10/094,257 (pending and published as US 2002-0170976). Briefly, as described therein, independent temperature control for each of the four or more reactors is effected using separately-controlled heating elements (e.g. resistive heating elements such as coil

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heaters) around each of the four or more reactors, while thermal isolation between the four or more reactors is accomplished by fluid-based heat exchange with an external heat sink. In preferred embodiments, the heat flux being applied to each of the reactors has an axial profile (taken along the length of the flow reactor, with the direction of flow) that can be varied (fixedly varied, or controllably varied) to compensate for variations in the heat-flux profile associated with the circulating heat-exchange fluid cooling the reactors, and to compensate for the varied locations of the four or more reactors relative to other reactors and to the external environment (*e.g.* centered reactors versus reactors near an external edge of the reactor module). Hence, design and/or control of the heating elements for each of the reactors can effect a substantially axially-uniform temperature profile for each of the four or more independently. Although especially useful in connection with parallel flow reactors, the temperature-control system disclosed in the aforementioned patent application can have applications for control of other types of reaction systems (*e.g.*, batch reactors, semi-continuous reactors) and/or in non-reaction chemical processing systems (*e.g.* calcining of heterogeneous catalysts) where parallel, independent temperature control is desirable.

[0126] A detection system especially preferred for use in connection with the reaction system of the present invention can comprise a multi-channel gas chromatograph as disclosed in co-owned, co-pending U.S. Serial. No. 60/222,540, entitled "Parallel Gas Chromatograph with Microdetector Array" filed August 2, 2000 by Srinivasan *et al.*, as well as in U.S. Serial No. 09/801,430, entitled "Parallel Gas Chromatograph with Microdetector Array" filed on March 7, 2001 by Srinivasan *et al.* (issued as U.S. Patent No. 6,701,774). Reactor effluents discharged from each of the four or more reactors can be simultaneously injected into such a multi-channel (*i.e.* parallel) gas chromatograph using a parallel injection valve, such as is disclosed in U.S. Serial No. 60/274,022, entitled "Gas Chromatograph Injection Valve Having Microvalve Array" filed on March 7, 2001 by Bergh *et al.*, ~~abandoned in favor of~~ filed as non-provisional applications U.S. Ser. No. 10/092,364 (pending and published as US 2002-0127146) and ~~in favor of~~ U.S. Ser. No. 10/092,035 (issued as U.S. Patent No. 6,742,544).

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[0146] The flow restrictor block 4510 also includes the six pairs of commonly-actuated inlet isolation microvalves 487, 489, as well as the outlet isolation valves 4580. These valves are preferably fabricated using precision machining techniques known in the art. Alternatively, the valves can be microfabricated, and can be integral with the flow-restrictor block 4510 or with a microchip body mounted thereon. The valves can also be, as noted above, part of an external fluid distribution system (480, Fig. 11b). The particular microvalve design is not critical. Preferably, the microvalves 487, 489 are membrane-actuated, membrane-seated valves such as shown in Figures 11H and 11I. Briefly, membrane-actuated valves 4300 can be prepared by precision machining to form the various component parts. In its open state (Fig. 11H), a fluid can flow into the valve through fluid inlet passage 4302, through internal passages 4303, past the valve seat 4310, and out through outlet passage 4304. In its closed state (Fig. 11I), a piston 4320 having a piston face 4322 is forced upward against a seating membrane 4315 such that fluid flow past the seat 4310 is sealingly blocked, with the seating membrane 4315 essentially acting as a gasket between the piston face 4322 and the valve seat 4310. The piston 4320 is preferably pneumatically actuated by use of an actuating membrane 4325 under pressure through actuation passage 4330. Portions of the seating membrane 4315 and actuating membrane that are situated between facing component surfaces of the valve body can serve as gaskets when the valve is clamped or fastened together. Further details are provided in co-pending, co-owned application U.S. Serial No. 60/274,022, entitled "Gas Chromatograph Injection Valve Having Microvalve Array" filed on March 7, 2001 by Bergh *et al.*, ~~abandoned in favor of~~ filed as non-provisional applications U.S. Ser. No. 10/092,364 (pending and published as US 2002-0127146) and ~~in favor of~~ U.S. Ser. No. 10/092,035 (issued as U.S. Patent No. 6,742,544).

[0148] The reactor module 4600, shown schematically in Figure 11O, comprises a 4x6 array of twenty-four reactor tubes 4610 individually supported in a reactor frame 4605. Each tube has a reaction volume of about 1 ml. Each of the reactor tubes 4610 can be individually heated using resistive coil heaters 4620 (*e.g.* Watlow Mini-K-ring). Thermal isolation between reactor tubes 4610 is achieved using fluid-type heat exchanger to cool the inter-reactor volume within the

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reactor frame 4610. Preferably, the cooling medium is air or inert gas, and is fed into the reactor module 4600 substantially at the midsection thereof. Plate cooling fluid (*e.g.* air) is also fed through the top member 4606 and bottom member 4607 of the reactor frame 4605, specifically through heat-exchange channels 4608 formed therein. Advantageously, as described in greater detail above, the heat flux associated with the resistive coil heaters 4620 can be axially varied to account for heat variations due to the reaction, and to balance heat removal by the cooling media such that a substantially axial uniform temperature profile is obtained. Further details about temperature control are provided in co-owned, co-pending application U.S. Serial No. 60/274,065, entitled "Parallel Flow Reactor Having Improved Thermal Control" filed on March 7, 2001 by Bergh *et al.*, ~~abandoned in favor of~~ filed as non-provisional application U.S. Ser. No. 10/094,257 (pending and published as US 2002-0170976). The feed gas flows into the reactor tube inlet 4612, and optionally contacts a catalyst (*e.g.*, supported in the reactor tube using frits (not shown)) under reaction conditions to effect the chemical reaction of interest. The reaction products and unreacted reactants are discharged through the reactor tube outlet 4614.

[NO FUTHER AMENDMENTS THIS PAGE]